

Magnetism of an adatom on biased AA-stacked bilayer graphene

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Abstract

We study magnetism of an adatom adsorbed on AA-stacked bilayer graphene (BLG) in both unbiased and biased cases, using the Anderson impurity model. We find different magnetic phase diagrams for the adatom, depending on the energy level of the adatom, which varies from the magnetic phase diagram of adatom in normal metals to that in graphene. This is due to the individual energy dependence of the density of states (DOS) of AA-stacked BLG and anomalous broadening of the adatom energy level. Furthermore we investigate the effect of a bias voltage on DOS of AA-stacked and show that the magnetization of the adatom can be controlled by applying the bias voltage. This allows for possibility of using AA-stacked BLG in spintronic devices.

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Keywords: AA-stacked bilayer graphene; Tight-binding model; Green's function; Magnetic phase diagram; Magnetization of adatom.

1 Introduction

Graphene is a single layer of carbon atoms arranged in a hexagonal lattice structure. Low-energy quasiparticles in graphene have linear dispersion relation and behave as massless Dirac particles [1]. This special dispersion relation leads to many unusual properties have not been observed before graphene isolation [2, 3]. Properties of few-layer graphene materials depend on the number of layers and the stacking order [2]. For example BLG in AB stacking has gapless quadratic dispersion relation and shows properties [4, 5] which are different from graphene and also from the ordinary two dimensional electron gas. Also AA-stacked BLG, a new stable stacking order of graphene [6, 7], has properties which are different from those of SLG and BLG. In the AA-stacked BLG each sublattice of the top layer is located directly above the same one of the bottom layer. The AA-stacked BLG has special band structure composed of two hole-doped and electron-doped graphene-like bands [8, 9]. Its band structure includes two decouple doped SLG bands which are shifted relative to each other. Due to this especial band structure the AA-stacked BLG shows attractive properties [8, 9, 10, 11, 12, 13] which have not been observed in the other two dimensional materials.

In two previous decade, some successful experimental efforts [14, 15] to control the position of an impurity adsorbed on a two-dimensional open surface by scanning tunnelling microscope have been reported. These motivated several groups to consider magnetization of adatoms adsorbed on garphene [16, 17, 18, 19, 20, 21] and AB-stacked BLG [22, 23, 24, 25]. They

showed that the magnetization of the adatom adsorbed on graphene and BLG is more possible than the magnetization of an adatom embedded in normal metals. Also they found that it can be controlled by an electric field via back gate [16]. For graphene, these features have been verified by an experimental research recently [26]. The AA-stacked BLG, which is a new open surface two dimensional material with especial low-energy DOS [8, 9], can be appealing to consider the adatom magnetization (Note that DOS plays key role in the magnetization of the adatom). Furthermore in this paper we show that the DOS of AA-stacked BLG can be manipulated by a vertical electric field (bias voltage). Hence one can control the magnetization of the adatom. This feature allows for high possible application of the AA-stacked BLG in the spintronics devices. In this paper, motivated by these facts, we consider the magnetization of an adatom adsorbed on biased AA-stacked BLG surface.

This paper is organized as follows. In Sec. II we present model Hamiltonian and details of our calculation. Also we consider the effect of the bias voltage on the DOS of AA-stacked BLG which has key effect on the magnetization of the adatom. Sec. III is devoted to our results and discussion. First we consider necessary conditions for the magnetization of the adatom in the unbiased case. Then we focus on the effect of the bias voltage on the magnetization of the adatom. Finally the paper is ended with the summary and conclusions.

2 Model Hamiltonian

We consider magnetization of an adatom, with a inner localized orbital, which is adsorbed on a sublattice of a AA-stacked BLG lattice. The Hamiltonian of this system can be written as

$$H_T = H_{BLG} + H_{ad} + H_V, \quad (1)$$

where H_{BLG} and H_{ad} are the Hamiltonian of the pure biased AA-stacked BLG lattice and the Hamiltonian of the adatom respectively. Also the hybridization of the localized orbital of the adatom with the conduction sea of biased AA-stacked BLG is reflected in H_V .

The momentum-dependent Hamiltonian of the biased AA-stacked BLG in the nearest-neighbor approximation is given by (we use units such that $\hbar = 1$)

$$\begin{aligned}
H_{BLG} = & -t \sum_{m=1}^2 \sum_{\mathbf{k}\sigma} [\phi(\mathbf{k}) a_{m\mathbf{k}\sigma}^\dagger b_{m\mathbf{k}\sigma} + \phi^*(\mathbf{k}) b_{m\mathbf{k}\sigma}^\dagger a_{m\mathbf{k}\sigma}] \\
& -V \sum_{m=1}^2 \sum_{\mathbf{k}\sigma} (-1)^m [a_{m\mathbf{k}\sigma}^\dagger a_{m\mathbf{k}\sigma} + b_{m\mathbf{k}\sigma}^\dagger b_{m\mathbf{k}\sigma}] \\
& + \gamma \sum_{\mathbf{k}\sigma} [a_{1\mathbf{k}\sigma}^\dagger b_{2\mathbf{k}\sigma} + b_{2\mathbf{k}\sigma}^\dagger a_{1\mathbf{k}\sigma}], \tag{2}
\end{aligned}$$

where $a_{m\mathbf{k}\sigma}^\dagger (a_{m\mathbf{k}\sigma})$ creates (annihilates) an electron with spin σ at A sublattice in m -th layer. $t \sim 3$ eV and $\gamma \sim 0.2$ eV [27, 28] present the nearest neighbor intralayer and the interlayer hopping energies respectively and the applied bias voltage is equal to $2V$. In eq. (5) $\phi(\mathbf{k}) = \sum_{i=1}^3 e^{i\mathbf{k}\cdot\mathbf{d}_i}$ where \mathbf{d}_i are the nearest neighbor vectors which have been shown in fig. 1. The corresponding energy bands are

$$\varepsilon_{\lambda,s} = s\sqrt{\gamma^2 + V^2} + \lambda\phi(\mathbf{k}), \tag{3}$$

where s refers to *bonding/antibonding* label and $\lambda = \pm 1$ are the electron-like and hole-like band indexes. One can expand $|\phi(\mathbf{k})|$ around Dirac points (\mathbf{K} or \mathbf{K}') for $|\mathbf{q}| \ll |\mathbf{K}|$ (where $\mathbf{k} = \mathbf{q} + \mathbf{K}$) to obtain the low energy bands of biased AA-stacked BLG. In this limit $|\phi(\mathbf{k})| = v_F q$ where $v_F = 3ta/2$ is Fermi velocity. Plots of the low energy bands for three values of bias voltage, $V=-\gamma$, $V=0.0$ and $V=+\gamma$ have been shown in fig. 2(a). We see that in the biased AA-stacked BLG the $s=+$ and $s=-$ bands are shifted by $+\sqrt{\gamma^2 + V^2}/\gamma$ ($-\sqrt{\gamma^2 + V^2}/\gamma$) with respect to the same band in the unbiased case. Local DOS at anyone of equivalent sublattices in the top layer

of biased AA-stacked BLG is

$$N(\omega) = \frac{1}{2D^2} \left[\frac{\Pi + V}{\Pi} |\omega + \Pi| + \frac{\Pi - V}{\Pi} |\omega - \Pi| \right] \theta(D - |\omega|), \quad (4)$$

where $\Pi = \sqrt{\gamma^2 + V^2}$, D is the high-energy cutoff of AA-stacked BLG bandwidth and $\theta(x)$ is the step function. Note that to obtain local DOS at sublattices of the bottom layer it is enough to replace V with $-V$. In fig. 2(b) we have plotted the local DOS at the top layer for three values of bias voltage, $V=-\gamma$, $V=0.0$ and $V=+\gamma$. As it is clear from eq. (5) and also from fig. 2(b) one can control the local DOS around the adatom level (so the magnetization of the adatom) via a bias voltage.

The adatom Hamiltonian, H_{ad} , is given by

$$H_{ad} = (\varepsilon_0 + V) \sum_{\sigma} f_{\sigma}^{\dagger} f_{\sigma} + U n_{\uparrow} n_{\downarrow}, \quad (5)$$

where $(\varepsilon_0 + V)$ is the energy level of the adatom shifted by the bias voltage (Let us suppose that the adatom is located near the top layer so they have same electric potential.) and U is the Coulomb energy for double occupancy of the adatom level. $n_{\sigma} = f_{\sigma}^{\dagger} f_{\sigma}$ is the occupation number of the adatom level and $f_{\sigma}^{\dagger} (f_{\sigma})$ operator creates (annihilates) an electron with spin σ at the adatom level. By using the mean field approximation, the two-body coulomb interaction term can be decoupled to the single-body interactions as $\sum_{\sigma} \langle n_{-\sigma} \rangle f_{\sigma}^{\dagger} f_{\sigma} - \langle n_{\uparrow} \rangle \langle n_{\downarrow} \rangle$. So we can rewrite the adatom Hamiltonian as $\sum_{\sigma} \varepsilon_{\sigma} f_{\sigma}^{\dagger} f_{\sigma}$, where $\varepsilon_{\sigma} = \varepsilon_0 - U \langle n_{-\sigma} \rangle$ is renormalized energy of the adatom level.

The hybridization of the localized orbital of the adatom with the conduction sea of AA-stacked BLG is given by

$$H_V = \frac{V_f}{\sqrt{N}} \sum_{\sigma \mathbf{k}} (f_{\sigma}^{\dagger} a_{1\mathbf{k}\sigma} + a_{1\mathbf{k}\sigma}^{\dagger} f_{\sigma}), \quad (6)$$

where V_f is the hybridization strength and the Hamiltonian has been written for an adatom adsorbed on an A sublattice at top layer and N is the number of A sublattices.

To determine the magnetization of the adatom one must obtain the occupation number of the spin up and down of the adatom level. If $n_{\uparrow}^{ad} \neq n_{\downarrow}^{ad}$, the adatom is magnetized and if they are equal, it isn't magnetized. The zero temperature occupation number of the adatom level for spin σ is given by

$$n_{\sigma}^{ad} = \int_{-\infty}^{\mu} d\omega \rho_{\sigma}^{ad}(\omega), \quad (7)$$

where $\rho_{\sigma}^{ad}(\omega) = -\frac{1}{\pi} \text{Im} G_{\sigma}^{ad,R}(\omega)$ and one can use the equation of motion technique to write $G_{\sigma}^{ad,R}(\omega)$ as

$$G_{\sigma}^{ad,R}(\omega) = [\omega - \varepsilon_{\sigma} - \Sigma^{ad,R}(\omega) + i0^+]^{-1}. \quad (8)$$

Here $\Sigma^{ad,R}(\omega)$ is the retarded self-energy and it can be written as

$$\Sigma^{ad,R}(\omega) = V_f^2 G_{\sigma}^{0R}(\omega) = \frac{V^2}{\sqrt{N}} \sum_{\mathbf{q}} G_{\sigma}^{0R}(\mathbf{q}, \omega), \quad (9)$$

where $G_{\sigma}^{0R}(\mathbf{q}, \omega)$ is the pure Green's function of biased AA-stacked BLG. After integration over the momentum we obtained a relation for the Green's functions as

$$\begin{aligned} G_{\sigma}^{0R}(\omega) &= -\frac{1}{2D^2} [(\omega - \Pi) \frac{\Pi + V}{\Pi} \ln \left| \frac{D^2 - (\omega - \Pi)^2}{(\omega - \Pi)^2} \right|] \\ &+ \frac{1}{2D^2} [(\omega + \Pi) \frac{\Pi - V}{\Pi} \ln \left| \frac{D^2 - (\omega + \Pi)^2}{(\omega + \Pi)^2} \right|] \\ &- i \frac{\pi}{2D^2} \left[\frac{\Pi + V}{\Pi} |\omega + \Pi| + \frac{\Pi - V}{\Pi} |\omega - \Pi| \right] \\ &\times \theta(D - |\omega|). \end{aligned} \quad (10)$$

One can solve eqs. (7)-(10) self-consistently to obtain the occupation number and determine the magnetization of the adatom. We present our numerical results in the next section.

3 Numerical results and discussion

Here we present our numerical results for magnetization of an adatom adsorbed on AA-stacked BLG. Firstly, we consider dependence of the adatom magnetic phase diagram on the adatom energy level for unbiased AA-stacked BLG (the adatom magnetic phase diagram separates the magnetic and nonmagnetic phase of the adatom). Secondly, we investigate the effect of the bias voltage on the magnetization of the adatom.

Unbiased AA-stacked BLG has special DOS, constant DOS (similar to normal metals) at low energies and linear DOS (similar to graphene) otherwise. This can lead to different magnetic phase diagrams for the adatom, depending on the energy level of the adatom. Figure 3 shows our results for plots of the boundary between the magnetic and nonmagnetic state of an adatom adsorbed on unbiased AA-stacked BLG, for three different values of the adatom energy level, $|\varepsilon_0| = 0.25\gamma$, $|\varepsilon_0| \sim \gamma$ and $|\varepsilon_0| = 2.0\gamma$. The left panels correspond to $\varepsilon_0 < 0$ and the right ones correspond to $\varepsilon_0 > 0$. When the energy level of the adatom approaches to zero (here for example $|\varepsilon_0| = 0.25\gamma$), the magnetic boundary becomes symmetric around the $y=0.5$ approximately and does not cross the $y=0.0$ (or $y=1.0$). This is similar to the phase diagram of a magnetic atom embedded in the normal metals[29]. This behavior can be understood by this fact that DOS of AA-stacked BLG around $\omega = 0$ is constant and nonzero (fig. 2 (b)). While for $\varepsilon_0 \sim +\gamma$ ($\varepsilon_0 \sim -\gamma$), the magnetic boundary crosses the line $y=0.0$ ($y=1.0$) namely the adatom can be magnetized even when the bare energy level is above (under) the Fermi level energy. This feature is due to the large broadening of the adatom energy level. If $|\varepsilon_0|$ increase more, the magnetic boundary becomes more asymmetric as the magnetic phase diagram of adatom in graphene. The large broadening of the adatom energy level (even larger than the

broadening of an adatom level with $|\varepsilon_0| \sim \gamma$), which is due to the linear energy dependence of DOS around the adatom energy level, is origin of this feature. Furthermore we see that for large $|\varepsilon_0|$ (larger than inter layer hopping) the magnetic area shrink in the $\pi V_f^2/DU$ direction, namely the adatom magnetization is possible only for larger values of U . This is due to the large amount of the DOS around the adatom energy level which enhances the hybridization of adatom energy level with conduction sea and so limits the magnetization of the adatom. We see that the adatom adsorbed on AA-stacked BLG shows different magnetic phase diagrams, depending on the energy level of the adatom, which varies from local moment phase diagram in normal metals to that in graphene. This is a new aspect not found in graphene and AB-stacked BLG.

Hereafter, we investigate the effect of an applied bias voltage on the magnetization of the adatom adsorbed on undoped AA-stacked BLG. As shown in Fig. 2 (b), DOS of AA-stacked BLG can be manipulate by applying the bias voltage. This allows to control the hybridization of the adatom energy level with the conduction sea of AA-stacked BLG and to control the magnetization of the adatom. This is possible for a wide range of the adatom energy levels and on site coulomb energies as shown in figs. 4 and 5. In fig. 4, plots of magnetization versus the bias voltage have been shown for different energy levels. We see that for a wide range of adatom energy levels, there is a bias voltage which can be applied to suppress the DOS around the adatom energy level. This decreases the hybridization of the adatom energy level with conduction sea and so the magnetization of the adatom. Fig. 5 shows plots of the magnetization of the adatom for different on site coulomb energies. We see that even at a small on site coulomb energy about 0.1 eV, the adatom is magnetized with a large amount of the magnetic moment about $0.7 \mu_B$ and its magnetization can be controlled by the bias voltage.

Controlling the magnetization of the adatom, specially for a such wide range of the adatom energy levels and the on site coulomb energies, allows for possibility of using AA-stacked BLG in spintronic devices.

4 Summary and conclusion

In summary, we applied the Anderson impurity model to study magnetization of an adatom adsorbed on AA-stacked BLG. We did our investigation for both biased and unbiased AA-stacked BLG. In the unbiased case, we found different magnetic phase diagrams for the adatom. We showed that this varies from magnetic phase diagram of an adatom embedded in normal metals to that for an adatom adsorbed on graphene surface, depending on the bare energy level of the adatom. We explained this feature based on the individual energy dependence of the DOS of AA-stacked BLG and anomalous broadenings of the adatom energy level. Furthermore we investigated the effect of an applied bias voltage on DOS of AA-stacked BLG and showed that one can control the magnetization of the adatom via the bias voltage for wide range of adatom energy levels and on site coulomb energies. This allows for application of AA-stacked BLG in spintronic devices.

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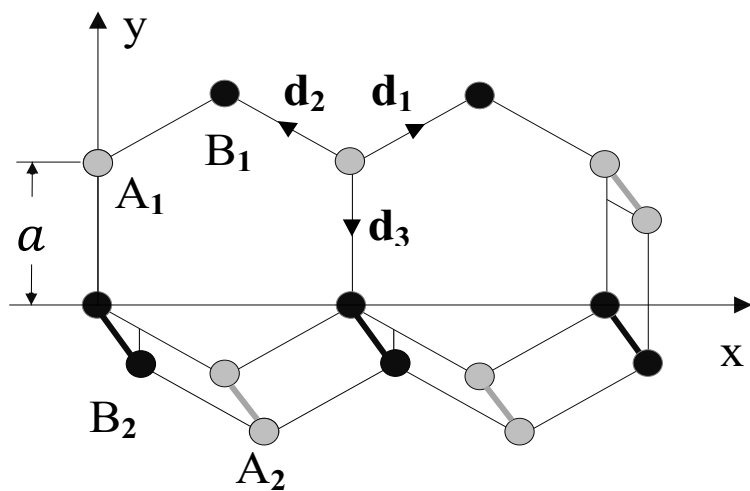


Figure 1: Lattice structure of AA-stacked BLG. $\mathbf{d}_1 = (a\sqrt{3}/2, a/2)$, $\mathbf{d}_2 = (-a\sqrt{3}/2, a/2)$ and $\mathbf{d}_3 = (0, -a)$ are three vectors that are drawn from connects A_1 sublattice to its nearest neighbors.

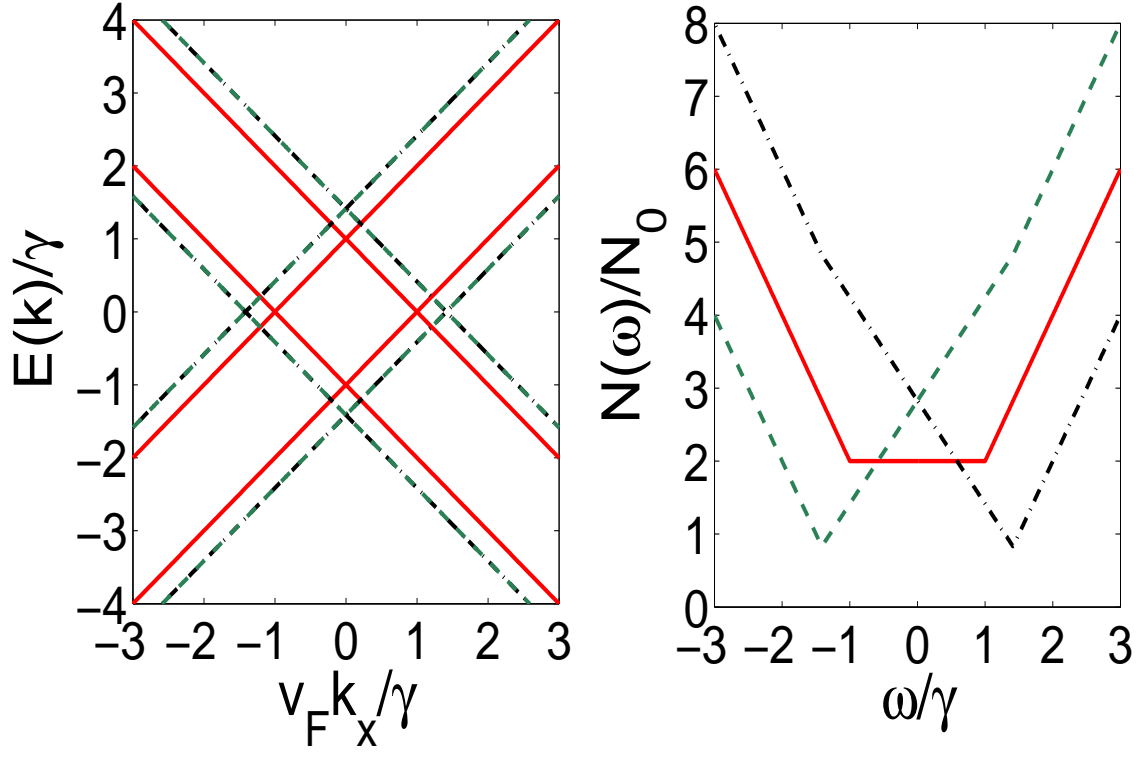


Figure 2: (a) shows the low-energy band structure of biased AA-stacked BLG for three values of the bias voltage $V = 0.0$ (solid line), $V = -\gamma$ (dashed line) and $V = +\gamma$ (dotted-dashed line) and (b) shows the DOS on a sublattice of top layer for these values of the bias voltage where $N_0 = 2\gamma/v_F^2$.

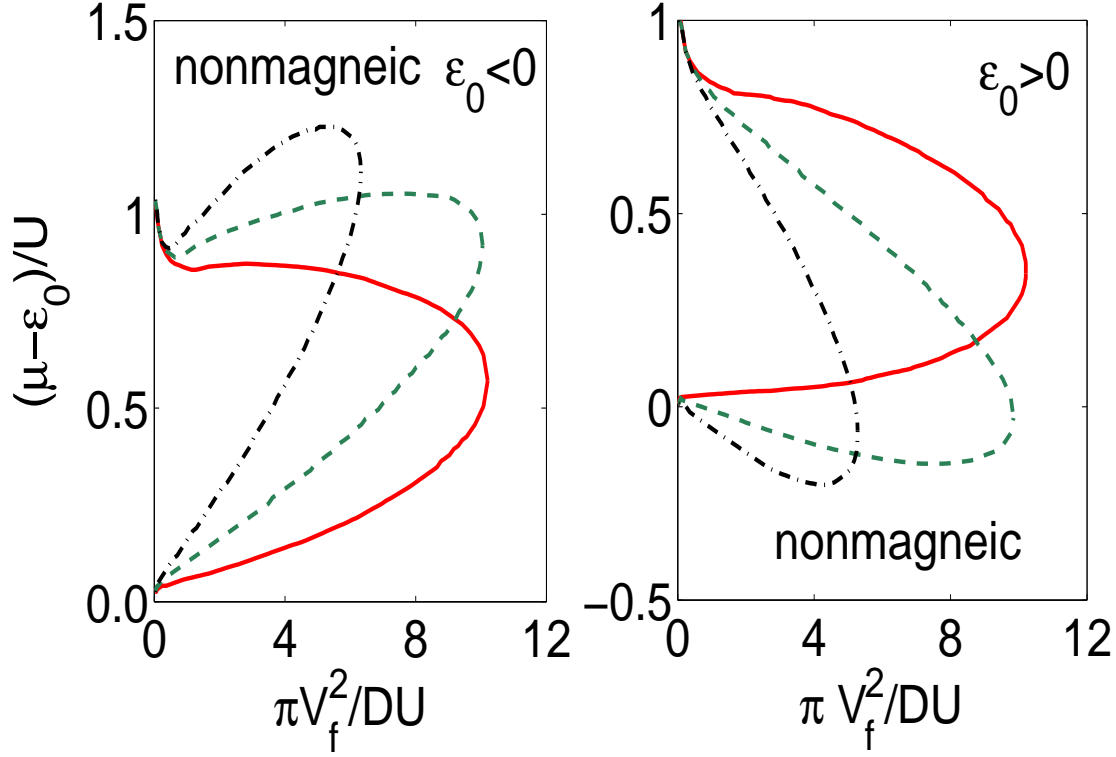


Figure 3: The boundary between magnetic and non-magnetic states of an adatom adsorbed on a sublattice of the unbiased AA-stacked BLG for three different values of the adatom energy level, $|\varepsilon_0| = 0.25\gamma$ (solid curves), $|\varepsilon_0| = 1.00\gamma$ (dashed curves) and $|\varepsilon_0| = 2.00\gamma$ (dotted-dashed curves). The right panels are for $\varepsilon_0 < 0$ and the left ones for $\varepsilon_0 > 0$. The other parameters are $\gamma = 0.2eV$, $V_f = 1eV$ and $D \sim 7eV$.

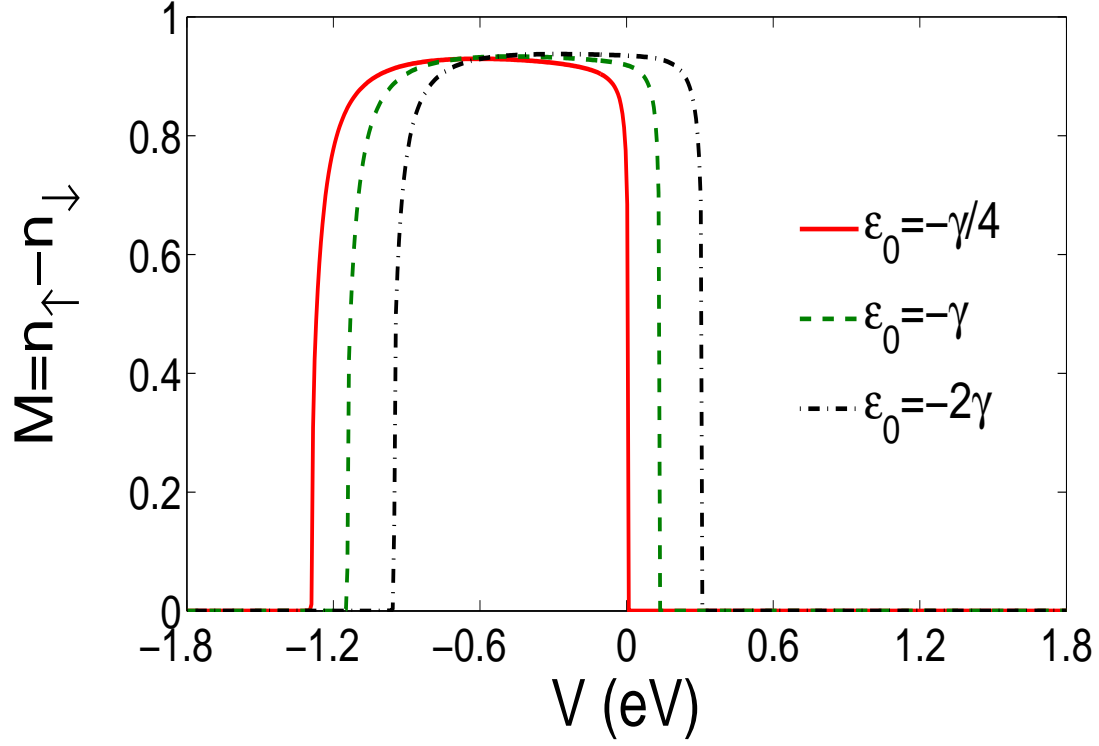


Figure 4: Plots of magnetization of the adatom vs. the applied bias voltage for different values of the adatom energy level, $\epsilon_0 = -\gamma/4$, $\epsilon_0 = -\gamma$ and $\epsilon_0 = -2\gamma$. The other parameters are $\gamma = 0.2\text{eV}$, $V_f = 1\text{eV}$, $U=1.5\text{ eV}$ and $\mu = 0$.

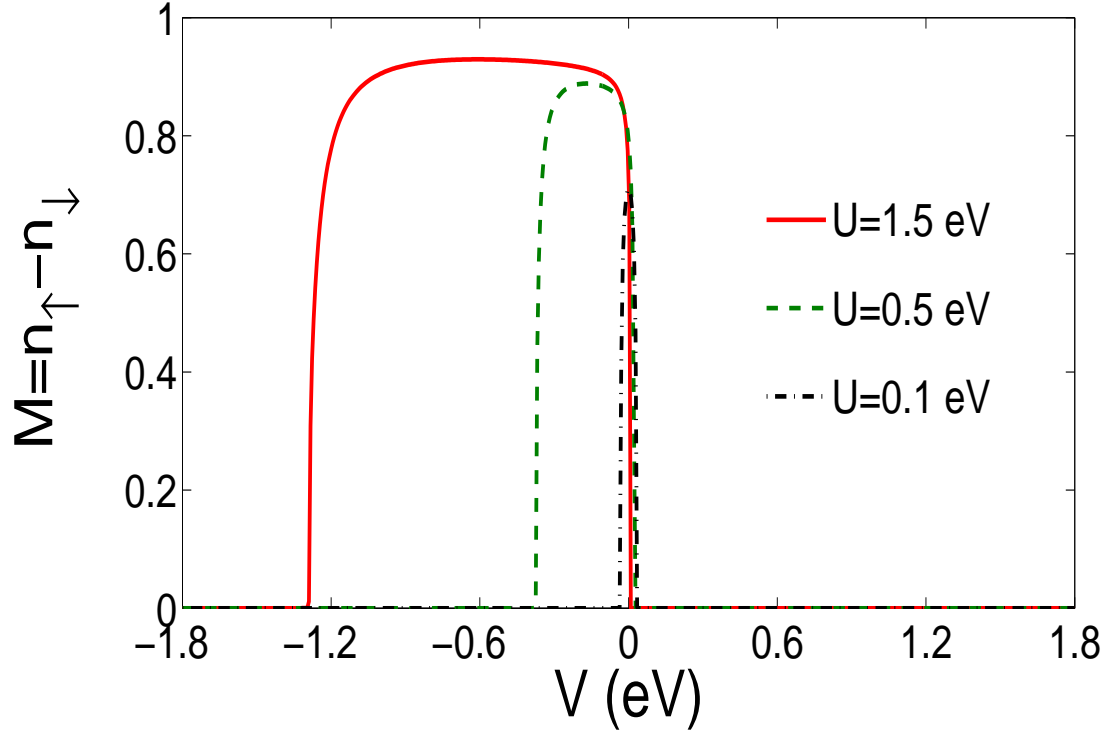


Figure 5: $M(V)$ of the adatom for different values of the on site coulomb energy, $U=1.5$ eV, $U=0.5$ eV and $U=0.1$ eV. The other parameters are $\gamma = 0.2eV$, $V_f = 1eV$, $\varepsilon_0 = -\gamma/4$ and $\mu = 0$.